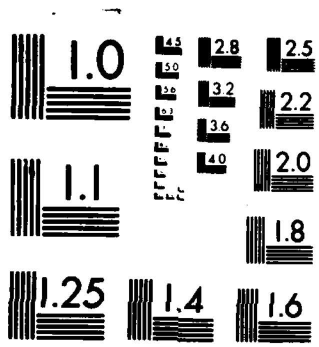
AD-A129 284 SPECTROSCOPIC INVESTIGATION OF LASER-INITIATED LOW-PRESSURE PLASMAS IN ATMOSPHERIC GASES(U) AIR FORCE GEOPHYSICS LAB HANSCOM AFB MA R A ARMSTRONG ET AL. 03 NOV 82 AFGL-TR-82-0367 1/1

UNCLASSIFIED F/G 20/5 NL

END
DATE
FILMED
7 83
DTIC



MICROCOPY RESOLUTION TEST CHART
NATIONAL BUREAU OF STANDARDS-1963-A

AFGL-TR-82-0367
PHYSICAL SCIENCES RESEARCH PAPERS, NO. 883

Spectroscopic Investigation of Laser-Initiated Low-Pressure Plasmas in Atmospheric Gases

R.A. ARMSTRONG
R.A. LUCHT
W.T. RAWLINS

3 November 1982

Approved for public release; distribution unlimited.

OPTICAL PHYSICS DIVISION PROJECT 2310
AIR FORCE GEOPHYSICS LABORATORY
HANSCOM AFB, MASSACHUSETTS 01731

AIR FORCE SYSTEMS COMMAND, USAF

This report has been reviewed by the ESD Public Affairs Office (PA) and is releasable to the National Technical Information Service (NTIS).

This technical report has been reviewed and is approved for publication.

Alva T. Stair, Jr.

DR. ALVA T. STAIR, Jr
Chief Scientist

Qualified requestors may obtain additional copies from the Defense Technical Information Center. All others should apply to the National Technical Information Service.

Unclassified

SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered)

REPORT DOCUMENTATION PAGE		READ INSTRUCTIONS BEFORE COMPLETING FORM
1. REPORT NUMBER AFGL-TR-82-0367	2. GOVT ACCESSION NO. AD-A125 284	3. RECIPIENT'S CATALOG NUMBER
4. TITLE (and Subtitle) SPECTROSCOPIC INVESTIGATION OF LASER-INITIATED LOW-PRESSURE PLASMAS IN ATMOSPHERIC GASES		5. TYPE OF REPORT & PERIOD COVERED Scientific. Interim.
		6. PERFORMING ORG. REPORT NUMBER PSRP, No. 663
7. AUTHOR(s) R.A. Armstrong, R.A. Lucht*, W.T. Rawlins**		8. CONTRACT OR GRANT NUMBER(s)
9. PERFORMING ORGANIZATION NAME AND ADDRESS Air Force Geophysics Laboratory (OPR-1) Hanscom AFB Massachusetts 01731		10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS 61102F 2310G414
11. CONTROLLING OFFICE NAME AND ADDRESS Air Force Geophysics Laboratory (OPR-1) Hanscom AFB Massachusetts 01731		12. REPORT DATE 3 November 1982
		13. NUMBER OF PAGES 15
14. MONITORING AGENCY NAME & ADDRESS(if different from Controlling Office)		15. SECURITY CLASS. (of this report) Unclassified
		15a. DECLASSIFICATION DOWNGRADING SCHEDULE

16. DISTRIBUTION STATEMENT (of this Report)

Approved for public release; distribution unlimited.

17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)

18. SUPPLEMENTARY NOTES

*Los Alamos National Laboratory
Group AP-3/M.S. 565
Los Alamos, NM 87545

**Physical Sciences, Inc.
Research Park
Andover, MA 01810

19. KEY WORDS (Continue on reverse side if necessary and identify by block number)

Laser
Plasma
Time resolved spectroscopy
Oxygen
Nitrogen
Argon
Multiphoton
Cascade

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

Laser-initiated breakdown of N_2, O_2, and Ar has been investigated at sub-atmospheric pressures using a pulsed (10 ns) Nd:yttrium aluminum garnet (Nd:YAG) laser at 1.06 μm. Breakdown thresholds have been determined from 3 to 760 torr and spectral analysis of the plasma emission has been carried out over the range 300-600 nm. Sharp spectral features arising from highly excited singly and multiply charged ions are reported, as well as an underlying continuum at intermediate pressure. Time resolved measurements indicate a complex kinetic dynamic optical behavior.

DD FORM 1 JAN 73 1473 EDITION OF 1 NOV 65 IS OBSOLETE

Unclassified

SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered)

Unclassified

Preface

This work was supported in part by the Air Force Geophysics Laboratory and Office of Scientific Research, and Defense Nuclear Agency. Additional technical contributions were provided by P. E. Nebolsine and G. E. Caledonia, Physical Sciences, Inc.

Contents

1. INTRODUCTION 7
2. PLASMA SPECTROSCOPY INVESTIGATIONS 8
3. CONCLUSIONS 12

REFERENCES 15

Illustrations

1. Apparent Breakdown Thresholds for Ar, N_2 and O_2 9
2. Temporal Profile of the 444.7 nm Line of N^+ 12

Spectroscopic Investigation of Laser-Initiated Low-Pressure Plasmas in Atmospheric Gases

I. INTRODUCTION

The subject of laser-initiated breakdown of gases has been of interest since the early application of lasers. Excellent articles have been published on some phases of this subject and Morgan[1] has reviewed the important results of the early work. The vast majority of the work has involved investigation of the energy coupling and breakdown threshold mechanisms, and resulting electron densities and temperatures. More recently, interest has increased in energy coupling at or near surfaces in support of laser fusion and laser damage studies. Relatively little attention, however, has been focused on understanding the time-resolved spectroscopy of the relaxing plasma after the breakdown. Stricker and Parker[2] have initiated some limited work in this area and Radziemski et al[3,4] have applied the technique to spectroscopic analysis of trace species. These investigations, however, are at relatively high pressures. Detailed knowledge of the spectroscopy and dynamics of plasmas of low pressure atmospheric gases is important for understanding the

(Received for publication 3 November 1982)

1. Morgan, C. Grey (1978) Sci. Prog. 65:31.
2. Stricker, J., and Parker, J.G. (1982) J. Appl. Phys. 53:851.
3. Radziemski, L.J., and Loree, T.R. (1981) Plasma Chemistry and Plasma Processing, 1(3):281.
4. Radziemski, L.J., Loree, T.R., Cremers, D.A., and Hoffman, N.M. unpublished.

behavior of upper atmospheric disturbances. The technique of laser-initiated breakdown can be used to simulate strong electron disturbances in the laboratory. We report here the results of initial investigations of the plasma spectroscopy, including additional information on breakdown thresholds, for Ar, N_2 and O_2 over the pressure range 3-760 torr, using focused laser radiation at 1.06 μm.

2. PLASMA SPECTROSCOPY INVESTIGATIONS

The experiments employed a Quanta-Ray DCR-1A Nd:yttrium aluminum garnet (Nd:YAG) laser, which can produce up to 750 mJ/pulse at 1.06 μm with a pulse width of 10 ns full width half-maximum (FWHM). The pulse shape was somewhat "spikey" in normal operation without an etalon in the oscillator cavity. However, the pulses were not mode-locked, so the breakdown threshold data could be interpreted in a straightforward manner. No discernible difference was found with a single mode-generating etalon inserted into the cavity. The unstable resonator cavity produced a 0.65 cm-(diameter) annular beam in the near field with a divergence θ < 0.5 mr (FWHM). The beam was focussed into the gas chamber by a short focal length lens (f = 5.8 cm) with less than one wavelength of spherical aberration. The laser was pulsed typically at 10 Hz; the results were not affected significantly by reducing the frequency to 2 Hz.

Apparent breakdown thresholds were determined by observing broadband (300-700 nm) emission from the focal volume. For pressures above 25 torr, the onset of breakdown was defined by the sudden appearance to the eye of visible emission from the focal volume. However, this method proved to be unreliable at pressures below 25 torr, so an RCA 1P28 photomultiplier tube was used to observe the plasma emission at lower pressures.

The results of the visual and photometric measurements for Ar, N_2 and O_2 are plotted in Figure 1. The data were fit to the function of $I_p = aP^b$, where I_p is peak intensity and P is the pressure in torr. The observed breakdown curves for all three gases are remarkably similar, with pressure exponents, b, of -0.3 to -0.5. An obvious change in slope occurs near 100 torr for argon and nitrogen, but not for oxygen. While the argon data are similar to Ireland's[5] for similar pulse widths, the break in slope was not evident from previous investigations using a Nd:YAG laser source.

---

5. Ireland, C. L. M. (1974) J. Phys. D: Appl. Phys. 7:L179.

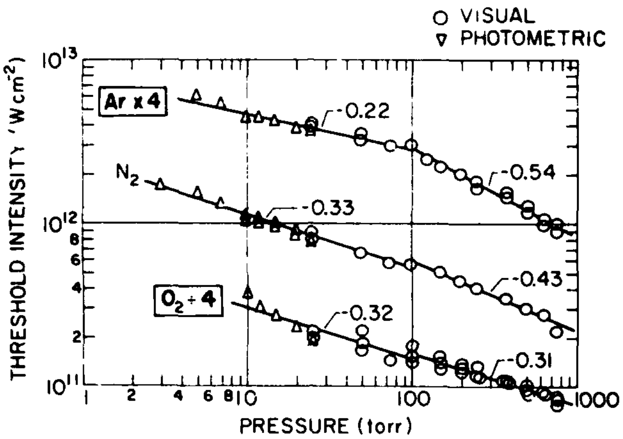


Figure 1. Apparent Breakdown Thresholds for Ar, N_2 and O_2. Pressure exponents, b, obtained from least squares fits (solid lines) for P > 100 torr and P < torr are indicated. For clarity, the argon and oxygen curves have been offset by a factor of 4 from their actual position

The pressure dependences above 100 torr are weaker than the P^{-1} behavior predicted by the theory of inverse bremsstrahlung "cascade",[1,6,7,8] in marked contrast to measurements for argon gas breakdown at 0.69 μm[9] which show a $\sim P^{-0.8}$ dependence. The weak pressure dependence in the present case may result from other effects such as multiple-photon inverse bremsstrahlung ionization,[10] or self-focussing, which have not been included in theoretical treatments.

At sufficiently low pressure, direct multiphoton ionization should dominate over the cascade mechanism,[1] resulting in a change of slope of the pressure dependence plots to $\sim P^{-1/n}$ where n is the number of photons required for multistep ionization. The previously unreported break in the slope of I_p vs P occurs at nearly an order of magnitude higher pressure than the theoretically predicted $P < 10^{-7}/\tau$ where τ is the laser pulse width in seconds. However, we do not observe an approach to $P^{-1/n}$ behavior as seen in cases of visible, picosecond timescale generated breakdown,[11] a result consistent with findings of other investigations.[1,5] This observation is difficult to understand for several reasons. First, below 10 torr, the electron mean free path is long relative to the spark dimensions. We calculate our focal radius and length to be 14.5 μm and 107 μm respectively, from which we derive an effective cylindrical electron diffusion length of 6 μm.

(Due to the large number of references cited above, they will not be listed here. See References, page 15.)

Since the electron mean free path in air is ~10 μm at 1 torr, and ~10 inverse bremsstrahlung events are required per electron to achieve ionization, collisional processes should not dominate at pressures below 10 μm. Second, our photometric results below 25 torr show a significant range of laser intensities near threshold over which plasma emission can be observed, similar to picosecond pulse width observations[11] which exhibited optical behavior consistent with multiphoton breakdown (~ 10^{-3} torr). Third, Baravian et al[12] have shown, using ion counting, that the multiphoton process dominates up to at least a few torr. Thus, although our low pressure data should reflect multiphoton controlled ionization, the optical behavior of the pressure dependence of breakdown intensity, I_p, is more consistent with cascade breakdown.

Spectra of the plasma emission following the laser pulse were obtained for the three gases from 300 to 600 nm. Gas pressures of 10 and 125 torr were used and the laser intensity was held at twice the apparent threshold value in each case. The spectra were obtained with a scanning monochromator (McPherson 219) and a boxcar integrator (PAR 162/165) with a 200 ns-wide gate, averaging over several laser pulses for each element of spectral resolution. The electronically excited species identified[13,14] from the spectra are N_2, N_2^+, N^+, N^{2+}, and N^{3+}, O^+ and O^{2+}, Ar^+ and Ar^{2+}, for breakdown in N_2, O_2, and Ar, respectively. The atomic states observed include high-lying Rydberg levels. Conspicuous by their absence are transitions of O_2, O_2^+, and O^{3+}. In similar experiments performed at pressures at and above 1 atm[2], the multiply charged species found here were not observed.

The nominal spectral resolution in our measurements is ~0.05 nm. The observed linewidths are 2-3 times broader; likely due to Stark broadening. Estimates of the electron density from the observed linewidths give ~ 10^{18} cm^{-3} at optically thin conditions of 10 torr, indicating the gas is essentially fully ionized, consistent with earlier investigations.[15] The prevalence of atomic transitions in the spectral data further indicates that the molecular gases are highly dissociated. In addition to the line spectra, a broad continuum was observed at 125 torr, but was absent at 10 torr. This time-integrated continuum, which is identical for O_2 and N_2, has a peak near 420 nm and an FWHM of about 150 nm, and is skewed to the red. The spectral distribution does not fit that of blackbody radiation, and we conclude that it is due to bremsstrahlung emission.

---

12. Baravian, G., Godart, J., and Sultan, G. (1980) Appl. Phys. Lett. **36**(6):415.

13. Pearse, R.W.B., and Gaydon, A.G. (1976) The Identification of Molecular Spectra, John Wiley and Sons, Inc., New York.

14. Wiese, W.L., Smith, M.W., and Glennon, B.M. (1966) Atomic Transition Probabilities, National Standard Reference Data System, National Bureau of Standard 4.

15. Braerman, W.F., Stumpfel, C.R., and Kunge, H.J. (1969) J. Appl. Phys. **40**(6):2549.

A measure of the excitation of N^+ and O^+ at a laser intensity of twice breakdown threshold was obtained from the spectra by plotting the observed relative emitting-state population, N_u, vs energy. Relative populations of N_u were determined from the observed peak intensities, I, of isolated atomic lines by $N_u \propto I\lambda/(A_{u1} g_u \epsilon_\lambda)$, where λ is the wavelength, ϵ_λ is the instrument spectral efficiency (as determined from separate calibrations), and A_{u1} and g_u are the upper level transition probability and degeneracy as obtained from compilations.[14] Apparent electronic "temperatures" were then determined by fitting the data to the Boltzmann formula $N_u \propto \exp(-E_u/kT)$, where E_u is the energy of the emitting species. These "temperatures" should be viewed with some caution since their accuracy is limited by that of the transition probabilities given in Reference 14. The results at 10 torr ($\pm 1\sigma$) give (17,900 ± 1,400)°K for N^+ and (26,700 ± 1,600)°K for O^+; at 125 torr, the apparent temperatures are (24,400 & 2700)°K for N^+ and (24,300 ± 2700)°K for O^+. The significant difference between the N^+ and O^+ temperatures at 10 torr reflects the larger energy required to excite high levels of N^+ from N_2. The similarity of the O^+ and N^+ temperatures at 125 torr may be accounted for if the plasma is optically thick. Estimates of the absorption coefficients for these transitions indicate that the plasma is indeed optically thick at 125 torr, but should be optically thin at 10 torr. The electronic temperature analysis does not assume any equilibrium conditions, but merely provides a measure of the extent of excitation of the observable states.

Time-resolved measurements were made of several emitting species with a 5 ns boxcar gate width. Nearly all visible emission occurs after the laser pulse. Measured species exhibited varied rise times of less than 100 ns at laser intensity of twice breakdown threshold for a given pressure, but in no case was peak emission reached within the laser pulse width. Emission decay rates of molecular nitrogen second positive and first negative systems were close to radiative decay rates. All atomic species, however, including those excited to high-lying energy levels, exhibited decay rates much longer than radiative. Species for which electronic temperatures were obtained exhibited constant relative population distribution throughout the decay. These results may be explained by a combination of spatial variations of source emitters and local thermodynamic equilibrium as presented by Radziemski.[4] However, increasing laser intensity results in nonuniform temporal relaxation as shown in Figure 2, a plot of the temporal profile of the 444.7 nm line of N^+. The upper curve exhibits a very complex kinetic and dynamic behavior which is not consistent with a thermodynamic equilibrium model.

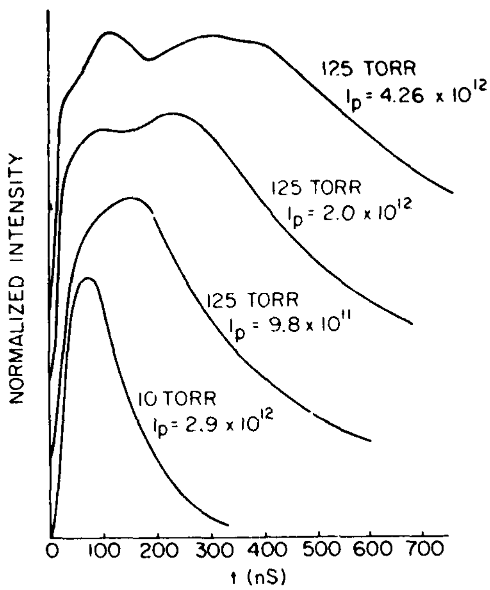


Figure 2. Temporal Profile of the 444.7 nm Line of N^+. I_p is peak laser intensity in W cm^{-2} at the pressures indicated. The 10 torr and first 125 torr curves are generated at I_p of twice threshold for breakdown

3. CONCLUSIONS

In conclusion, although several articles on laser induced gas breakdown appeared in the 1960's literature, an adequate explanation of the failure of nano-second pulse width 1.0 μm laser breakdown to show clear crossover to multi-photon behavior at low pressure has never emerged. Some recent articles purport to explain the behavior but they fail to fit the data, and cannot be generally applied to all laser breakdown conditions. We have used a photometric scheme to determine the low pressure breakdown thresholds and by so doing, identified a heretofore unreported break in the slope of breakdown threshold vs pressure for a 10 ns pulse width Nd:YAG laser. We have also reported new data on O_2 and N_2 as well as time behavior which is not explainable by a thermal equilibrium model.

While we do not yet have a full explanation of the optical behavior of the laser produced plasma, it is clear that simple models of multiphoton vs cascade processes are not adequate. We also caution that it is difficult and somewhat arbitrary to assign breakdown "threshold" at reduced pressures and the onset of detectable emission from the focal volume may be an insufficient criterion for the attainment

of breakdown "threshold". The issues cannot be resolved without a detailed experimental and theoretical investigation of the coupled gas dynamics, optics and kinetics of the laser initiated plasma. We are continuing to investigate these processes.

References

1. Morgan, C. Grey (1978) Sci. Prog. 65:31
2. Stricker, J., and Parker, J.G. (1982) J. Appl. Phys. 53:851.
3. Radziemski, L.J., and Loree, T.R. (1981) Plasma Chemistry and Plasma Processing, 1(3):281.
4. Radziemski, L.J., Loree, T.R., Cremers, D.A., and Hoffman, N.M. unpublished.
5. Ireland, C.L.M. (1974) J. Phys. D: Appl. Phys. 7:L179.
6. Ireland, C.L.M., and Morgan, C. Grey (1973) J. Phys. D: Appl. Phys. 6:720.
7. Morgan, F., Evans, L.R., and Morgan, C. Grey (1971) J. Phys. D: Appl. Phys. 4:225.
8. Gamal, Y., and Harith, M. (1981) J. Phys. D: Appl. Phys. 14:2209.
9. Dewhurst, R.J. (1977) J. Phys. D: Appl. Phys. 10:283.
10. Dewhurst, R.J. (1978) J. Phys. D: Appl. Phys. 11:L191.
11. Agostini, P., Barjot, G., Mainfray, G., Manus, C., and Thebault, J. (1970) IEEE J. Quantum Elect. 6:782.
12. Baravian, G., Godart, J., and Sultan, G. (1980) Appl. Phys. Lett. 36(6):415.
13. Pearse, R.W.B., and Gaydon, A.G. (1976) The Identification of Molecular Spectra, John Wiley and Sons, Inc., New York.
14. Wiese, W.L., Smith, M.W., and Glennon, B.M. (1966) Atomic Transition Probabilities, National Standard Reference Data System, National Bureau of Standard 4.
15. Braerman, W.F., Stumpfel, C.R., and Kunge, H.J. (1969) J. Appl. Phys. 40(6):2549.